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Theoretical Evaluation of Physical and Radiation Attenuation Properties of Yttrium Borate Glasses Modified by Tantalum Oxide

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Abstract

The physical and gamma-ray attenuation parameters of the free lead glass system xTa_2O_5–$20Yb_2O_3$–$(80–x)B_2O_3$ (x = 10, 20, 30, and 40 mol%) were theoretically investigated. The analyzed physical properties included empirical density (ρ_{emp}), molar volume (V_M), oxygen molar volume (V_O), oxygen packing density (OPD), and Poisson's ratio (σ). Data analysis showed that the ρ_{emp}, V_M, and V_O increased with higher Ta_2O_5 content. A corresponding decrease in OPD accompanied this increase. All samples studied maintained a constant σ of 0.24, indicating stable elastic behavior despite variations in glass composition. Furthermore, shielding parameters were theoretically calculated using WinXCom software at photon energies of 0.662, 1.173, and 1.33 MeV. The evaluated coefficients are mass attenuation coefficient (μ_m), linear attenuation coefficient (μ_L), half-value layer (HVL), tenth-value layer (TVL), and mean free path (MFP). The results indicated that LAC and MAC increased

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with higher Ta_2O_5 content, whereas HVL, TVL, and MFP decreased. The BT4 glass sample exhibited superior gamma-ray shielding performance compared with the other investigated compositions.

Keywords: Yttrium borate glasses; Tantalum oxide; Lead-free radiation shielding; Gamma-ray attenuation; Physical properties; WinXCom software; Half-value layer (HVL).

التقويم النظري للخصائص الفيزيائية ومعاملات توهين الإشعاع في زجاج بورات الإيتريوم المُعدَّل بأكسيد التنتالوم

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مّلخص:

تمت دراسة الخصائص الفيزيائية ومعاملات توهين أشعة غاما لنظام زجاجي خالٍ من الرصاص، يتكون من المركبات $xTa_2O_5–20Yb_2O_3–(80–x)B_2O_3$ (x = 10, 20, 30، 40 % موليا)، وذلك من الناحية النظرية. شملت الخصائص الفيزيائية التي تم تحليلها: الكثافة التجريبية(ρ_{emp})، والحجم المولي (V_M)، والحجم المولي للأكسجين (V_O)، وكثافة تعبئة الأكسجين (OPD)، ونسبة بواسون (σ). أظهر تحليل البيانات أن كلاً من (ρ_{emp}) و (V_M) و(V_O) تزداد بزيادة محتوى Ta_2O_5 ، بينما صاحب ذلك انخفاض في كثافة تعبئة الأكسجين OPD. وقد حافظت جميع العينات المدروسة على قيمة ثابتة لنسبة

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بواسون (σ) بلغت 0.24، مما يشير إلى استقرار السلوك المرن للمادة رغم التغيرات في تركيب الزجاج. علاوة على ذلك، تم حساب معاملات الحماية نظرياً باستخدام برنامج WinXCom عند طاقات فوتونية قدرها 0.662 و1.173 و1.33 ميغا إلكترون فولت (MeV). وشملت المعاملات التي تم تقييمها: معامل التوهين الكتلي (μ_m)، ومعامل التوهين الخطي (μ_L)، وطبقة القيمة النصفية (HVL)، وطبقة القيمة العشرية (TVL)، ومتوسط المسار الحر (MFP). وأظهرت النتائج أن كلاً من المعاملين LAC وMAC يزدادان مع زيادة محتوى Ta_2O_5، في حين تنخفض قيم كل من HVL وTVL وMFP. وقد أظهرت العينة الزجاجية BT4 أداءً فائقاً في الحماية من أشعة غاما مقارنة بالتركيبات الأخرى التي خضعت للدراسة.

الكلمات المفتاحية: زجاج بورات الإيتريوم؛ أكسيد التنتالوم؛ درع إشعاعي خالٍ من الرصاص؛ توهين أشعة غاما؛ الخصائص الفيزيائية؛ برنامج WinXCom؛ طبقة القيمة النصفية (HVL).

1. Introduction:

Over the past decades, the urgent need for developing new, safe, and environmentally friendly radiation shielding materials [1,2] has grown with the recent consistent increase in the application of radiation in various fields. Such materials are essential for protecting the environment from radiation exposure risks, enhancing public safety in nuclear and technological industries, improving health outcomes, and safeguarding human lives against the harmful effects of ionizing radiation. In particular, glasses with properties [3–5] such as transparency to visible light, ease of fabrication, and tunable characteristics through compositional and processing adjustments are increasingly employed as host materials for diverse shielding applications. For example, borate glasses are widely used in scientific and technological contexts due to their favorable properties, including low melting temperatures, high permeability, and excellent thermal stability in the ultraviolet (UV) and infrared (IR) regions. Moreover, glasses incorporating rare earth oxides (REE), such as yttrium oxide, have garnered significant attention due to their superior physical and optical characteristics,

including high refractive index, density, visible light transmittance, and thermal expansion as well as excellent infrared transmission [6,7]. The advantages of yttrium borate glasses make them highly desirable for applications, particularly in electro-optic devices, thermal and mechanical sensors, reflective windows, UV-blocking and attenuation shielding [6, 8]. Furthermore, the addition of such an oxide has been found to increase the effective atomic number and density, thereby improving radiation shielding performance. Considering these benefits, yttrium borate glasses have been identified as suitable host materials in diverse technological and biological applications. The unique features of transition metal oxide (TMO) glasses [7, 9], along with their potential suitability for advanced radiation applications, continue to attract significant research interest. One of the most significant transition metal oxides is tantalum oxide (Ta_2O_5), which possesses extraordinary physical and chemical properties [10, 11], including high dielectric and refractive coefficients, as well as excellent photoelectric performance. Ta_2O_5-doped glasses with high refractive indices are widely employed in optical applications and in the fabrication of ceramic capacitors and piezoelectric ceramics [12]. In addition, the lead-doped glass is an efficient transparent shield, but Pb toxicity and leaching hazards make lead-free glasses the safer option [13, 14]. Ta_2O_3 (Ta, Z = 73) increases density, refractive index and photon attenuation in silicate and tellurite glasses while keeping good transparency [15,16]. Likewise, Yb_2O_3 (Yb, Z = 70) raises density, mass attenuation coefficient and Z_{eff} and lowers the half-value layer in lead-free borate glasses, outperforming other heavy rare-earth oxides [17, 18]. Both are thus promising PbO replacements. Therefore, the primary objective of the present work is to carry out the investigation of yttrium borate glass containing tantalum oxide and figure out the possibilities of these systems becoming good candidates for gamma-shielding materials. In this study, theoretical evaluations were performed using the WinXCom software. The process involved the calculation of some gamma radiation shielding characteristics such as linear attenuation coefficient (μ), mass attenuation coefficient (MAC), half value layer (HVL), tenth value layer (TVL), and mean free path (MFP).

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2. Materials and Methods:

The selected glass samples in this research with compositions of xTa_2O_5-$20Yb_2O_3$-$(80-x)B_2O_3$ ($10 \leq x \leq 40$) (x in mol %) are presented in Table 1.

Table 1. Chemical compositions of the studied glasses

Sample Code	Chemical composition (mol%)		
	Ta_2O_5	Yb_2O_3	B_2O_3
TB1	10	20	70
TB2	20	20	60
TB3	30	20	50
TB4	40	20	40

2.1 Theoretical background

2.1.1 Physical parameters

The ρ_{emp} of the investigated glass samples is estimated using the following empirical equation [19]:

$$\rho_{emp} = 0.53 \frac{\sum M_i x_i}{\sum V_i x_i} \quad (1)$$

Where M_i, xi, and V_i are the molecular weight (Kg mol-1), the mole fraction (mole %), and the packing density parameter (m^3/mol) of the ith oxide component.

The packing density parameter (V_i) for M_XO_Y is calculated using the formula [19]:

$$V_i = \frac{4}{3}\pi N_A (x.r_M^3 + y.r_O^3) \quad (2)$$

Where r_M and r_O are the ionic radii of metal and oxygen, respectively.

The V_M of the glass matrix is defined the physical volume occupied by one mole of the glass network structural units and is given by [20]:

$$V_M = \frac{M}{\rho_{emp}} = \frac{\sum x_i M_i}{\rho_{emp}} \quad (3)$$

Where; M is the total molecular weight of glass mixture.

The V_O is defined as the volume of glass containing one mole of oxygen and is expressed in cm^3/mol. V_O is calculated using the following relation [20]:

$$V_O = \frac{V_M}{\sum x_i n_i} \quad (4)$$

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Where; n_i is the number of oxygen atoms in each component oxide. The OPD is a crucial factor in understanding the structure of glass, as it provides details on the density of the glass network, and it can be computed using the following equation [20]:

$$OPD = 1000C\left(\frac{\rho_{emp}}{M}\right) \quad (5)$$

Where C is the total number of oxygen atoms.

The total packing density (V_t) is a dimensionless structure factor representing the fraction of space occupied by the constituent ions relative to the glass matrix's total molar volume. It can be calculated by [20]:

$$V_t = \frac{\Sigma V_i X_i}{V_M} \quad (6)$$

Poisson's ratio (σ) is the ratio of lateral to longitudinal strain caused by a tensile force and can be expressed as follows [20]:

$$\sigma = 0.5 - \left(\frac{1}{7.2V_t}\right) \quad (7)$$

2.2.2 Radiation Attenuation Parameters

Using the WinXcom software, the radiation attenuation coefficients of tantalum–yttrium borate glasses with varying concentrations at photon energies of 0.662, 1.173, and 1.33 MeV were theoretically analyzed. WinXCom simulation software was developed to calculate photon interactions for any element, compound, or mixture across energies ranging from 1 keV to 10 GeV [21]. The program provides outputs including total attenuation with scattering, photoelectric absorption, pair production, and electron field interactions. The calculations of μ_m coefficient is based on Beer-Lambert law[11]:

$$I = I_0 e^{-\mu_m x} \quad (8)$$

Where, I is attenuated radiation, I_0 is the original intensity of radiation, and x is the thickness of the glass sample. In addition to μ_m, the key parameters under consideration for radiation shielding are μ_L, HVL, TVL, and MFP. μ_L describes the fraction of incident photons in a mono-energetic beam that are attenuated per unit thickness of a material. It includes all possible interactions, Compton scattering, the photoelectric effect, and pair production. The formulas for the parameters under investigation are obtained as follows [3-5]:

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$$\mu_{L(total)} = \mu_{photoelectric} + \mu_{Compton} + \mu_{pair\ production} \quad (9)$$

$$\mu_m = \sum_i W_i \left(\frac{\mu_{Li}}{\rho}\right) \quad (10)$$

Where μ_{Li} is the linear attenuation coefficient of the i^{th} element, ρ is the density of the sample, and W_i is the weight fraction of the i^{th} element in the glass sample.

Consequently, μ_L is calculated by the following equation.

$$\mu = \mu_m \rho \quad (11)$$

The HVL is the thickness of a material required to reduce the radiation intensity to 50% of the initial value. The HVL is calculated using the following equation:

$$HVL = \frac{\ln 2}{\mu_L} = \frac{0.693}{\mu_L} \quad (12)$$

The TVL is defined as the thickness needed to reduce radiation intensity to one-tenth of its original value and can be computed by:

$$TVL = \frac{\ln 10}{\mu_L} = \frac{2.302}{\mu_L} \quad (13)$$

The mean free path (MFP) explains how much radiation is absorbed or scattered per unit length of the material and can be calculated by the relation:

$$MFP = \frac{1}{\mu_L} \quad (14)$$

3. Results and Discussion

3.1. Physical Properties

The calculated parameters of ρ_{emp}, V_M, V_O, OPD, and σ by equations (1 – 7) are listed in Table 2 and graphically illustrated in Figures 1–3. As seen from this table and figures, the empirical density increased from 3.964 g cm^{-3} to 5.464 g cm^{-3} with the substitution of B_2O_3 by Ta_2O_5 in the glass structure. This increase in density can be attributed, in part, to the replacement of the relatively lighter B_2O_3 with the heavier Ta_2O_5, as the molecular weight of Ta_2O_5 (M_w= 441.893 g mol^{-1}) is considerably higher than that of B_2O_3 (M_w= 69.6182 g mol^{-1}), resulting in more glass density. The values of V_M increase from 43.31 cm^3/mol to 51.86 cm^3/mol as tantalum oxide content in the glass matrix increases. This indicates

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a possible expansion of the glass network, leading to a more open structure in the glass matrix [22]. This can be attributed to the fact that Ta_2O_5 produces more non-bridging oxygen in the glass matrix than B_2O_3.

Table 2. Physical parameters of Ta_2O_5-Yb_2O_3-B_2O_3 glasses.

Sample Code	ρemp (g/cm3)	M (g/mol)	VM (cm3/mol)	VO (cm3/mol)	OPD (mol/L)
TB1	3.964	171.70	43.31	13.53	73.89
TB2	4.526	208.92	46.16	13.58	73.66
TB3	5.022	246.14	49.01	13.61	73.46
TB4	5.464	283.36	51.86	13.65	73.28

The oxygen molar volume ranges between 13.53 and 13.65 cm^3/mol, while the oxygen packing density varies from 73.89 to 73.28 mol/L depending on the composition. The general tendency is that the OPD decreases slightly with the increase of Ta_2O_5 content in the glass composition. The replacement of B^{3+} ions (field strength: 0.71) with Ta^{5+} ions (field strength: 0.19) results in a higher molar volume of oxygen and a lower density of oxygen packing, i.e., a less compact glass network. At higher Ta_2O_5 concentrations, the decrease of the oxygen packing density is explained by the formation of non-bridging oxygens (NBOs), which results in an increase of the proportion of tetrahedral borate and yttrium groups and a decrease of the proportion of TaO_6 structural units. Thus, the replacement of the glass former B_2O_3 by Ta_2O_5 (as an intermediate) results in a higher proportion of non-bridging oxygen species over bridging ones, thus decreasing the overall connectivity of the modified borate glass network [23]. This observation highlights that the glass network expands with the formation of new structural units as Ta_2O_5 content increases, which in turn leads to an increase in molar volume.

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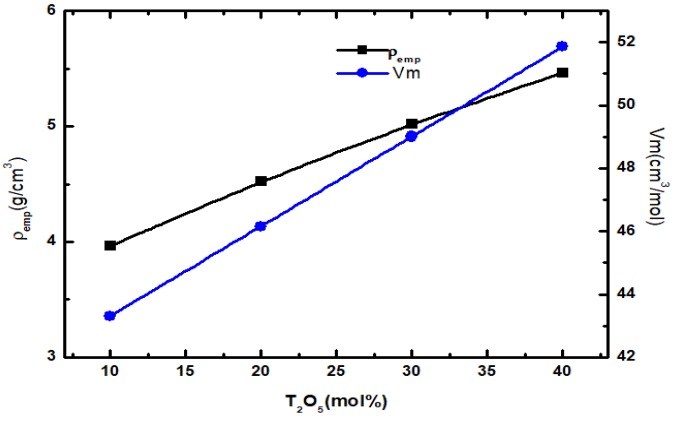


Fig.1: ρ_{emp} and V_M values as a function of T_2O_5 content mole% for Ta_2O_5-Yb_2O_3-B_2O_3 glasses.

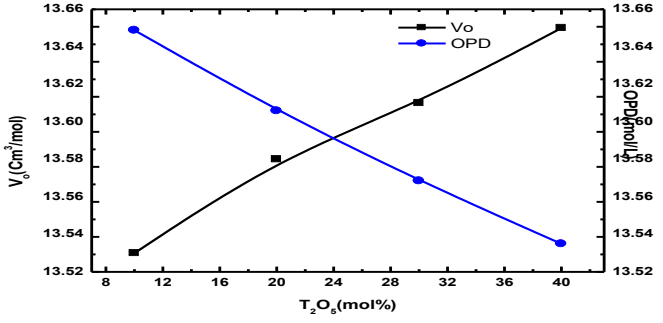


Fig.2: V_O and OPD values as functions of T_2O_5 content mole% for Ta_2O_5-Yb_2O_3-B_2O_3 glasses.

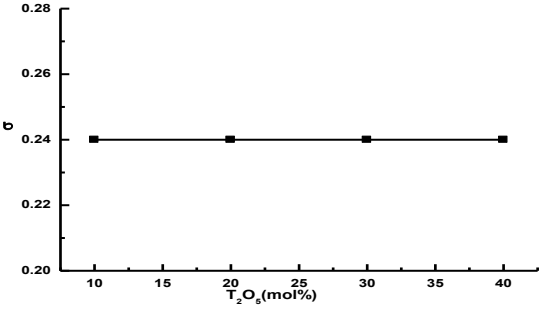


Fig.3: Variation of σ as a function of T_2O_5 content mole% for Ta_2O_5-Yb_2O_3-B_2O_3 glasses.

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σ represents the stiffness of the glass system. It is typically associated with mechanical qualities such as hardness and strength, with values ranging from 0 to 0.5. Materials with Poisson's ratio values less than 0.3 are generally considered to have a high cross-linking density, whilst those with values above 0.3 have a lower cross-linking density [24]. Figure 3, indicates that the prepared glass samples exhibit a nearly constant Poisson's ratio of approximately 0.24. This demonstrated that the studied glasses possess a high density of cross-links, making them suitable candidates for various important applications, particularly in radiation shielding.

3.2 Radiation Shielding Parameters

The radiation parameters μm, μL, HVL, TVL, and MFP for the examined xTa_2O_5–$20Yb_2O_3$–$(80 – x)B_2O_3$ (x = 10, 20, 30, 40 mol%) glass samples at multiple photon energies, 0.662, 1.173, and 1.331 MeV, are estimated theoretically using WinXcom software and presented in Tables 3 - 5.

Table 3. μ_m and μ_L parameters of the studied glasses.

Sample Code	ρ_{emp} g/cm	Photon Energy (MeV)					
		μ_m x 10^{-2} (cm^2g^{-1})			μ_L x 10^{-1} (cm^{-1})		
		0.662	1.173	1.332	0.662	1.173	1.332
TB1	3.964	8.66	5.72	5.30	3.43	2.27	2.10
TB2	4.526	8.85	5.76	5.33	4.01	2.61	2.41
TB3	5.022	8.91	5.77	5.33	4.47	2.90	2.68
TB4	5.464	9.06	5.78	5.33	4.95	3.16	2.91

Table 4. HVL and TVL parameters of the studied glasses.

Sample Code	Photon Energy (MeV)					
	HVL (cm)			TVL (cm)		
	0.662	1.173	1.332	0.662	1.173	1.332
TB1	2.019	3.057	3.299	6.707	10.154	10.959
TB2	1.730	2.659	2.873	5.748	8.832	9.545
TB3	1.549	2.392	2.589	5.145	7.946	8.602
TB4	1.400	2.195	2.380	4.651	7.291	7.906

Table 5. MFP parameter of the studied glasses.

Sample Code	Photon Energy (MeV)		
	MFP (cm)		
	0.662	1.173	1.332
TB1	2.91	4.41	4.76

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TB2	2.50	3.84	4.15
TB3	2.23	3.45	3.74
TB4	2.02	3.17	3.43

As seen in Table 3 and Figure 4, the μL values increased markedly as Ta_2O_5, increased whereas the μm increased slightly with increasing Ta_2O_5 content in the glass for each energy value increased. This is because the density of the glass samples increases noticeably as the Ta_2O_5 content in the glass network increases. The observed effect was more dominant at the lower energy (0.6MeV) compared with other chosen energy regions, since low-energy gamma rays have a lower penetration capability than medium- and high-energy photons.

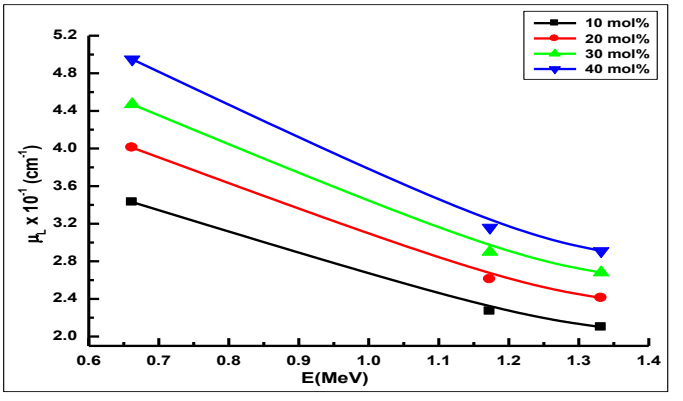


Fig.4: μ_L as a function of photon energy for the studied glasses.

Figures 5 illustrates the μ_m variation of the present glass samples versus the photon energies. The three different regions of lower, intermediate, and higher gamma energy were selected, allowing for a comprehensive understanding of the attenuation performance of glass samples across different energy levels. As it is notable, a sharp decrease in both values for all glass compositions is seen in both figures. This indicates that at lower photon energy, more photons are being attenuated, which might be related to the photoelectric effect interaction between gamma ray photons and the glass sample's atoms [25]. The BT4 glass sample with a 40% Ta_2O_5 content exhibited the highest $\mu_{L\ and}\ \mu_m$ values at all studied energies compared to the other samples, as seen in Table 3.

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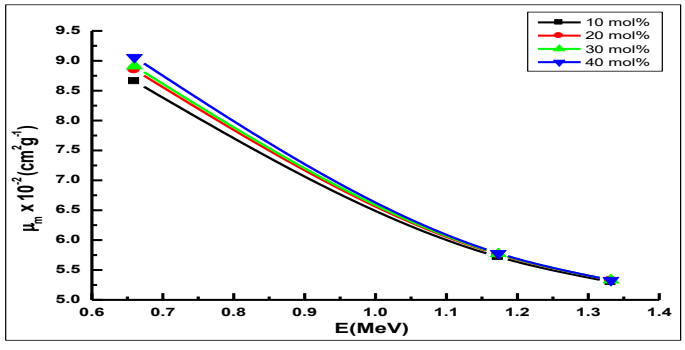


Fig.5: μ_m as a function of photon energy for the studied glasses.

HVL and TVL are important parameters for evaluating the penetration of radiation through shielding materials and assessing their shielding effectiveness. HVL is the thickness of material that will decrease radiation intensity to 50% of the initial value, while TVL is the thickness of material required to reduce the radiation intensity to 10% of the original beam [26]. The calculated HVL and TVL values for all glasses at given photon energies are listed in Table 4 and plotted in Figures 6 and 7. As observed, the HVL and TVL values are lowest at lower photon energies and increase with increasing photon energy. This indicates that high-energy photons require a greater thickness of the glass material to achieve the same attenuation level and, consequently, have a greater penetration capability than low-energy photons.

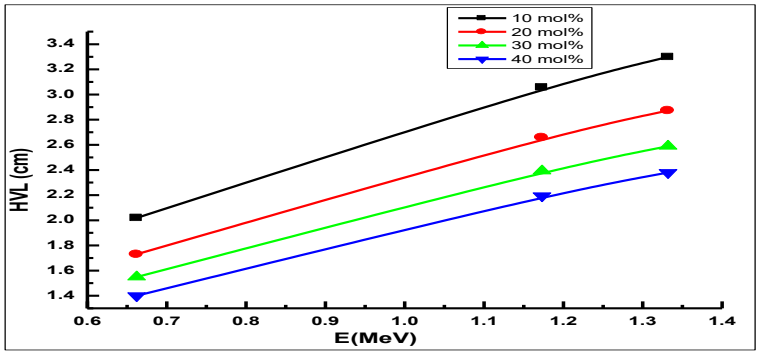


Fig.6: HVL as a function of photon energy for the studied glasses.

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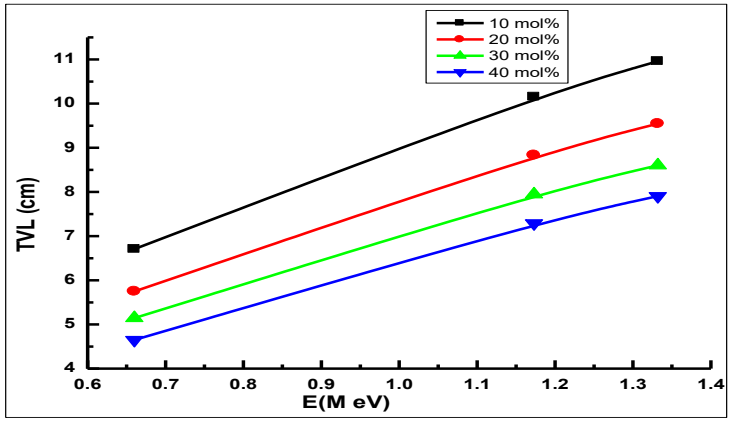


Fig.7: TVL as a function of photon energy for the studied glasses.

Furthermore, the data shown in Table 4 and presented in Figures 8 and 9 indicated that both HVL and TVL values decreased with the introduction of Ta_2O_5 content. One noticed that the addition of Ta_2O_5 led to an increase in the density of the sample, and it is well known that as the density becomes higher, the more effective radiation shielding will be enhanced [27].

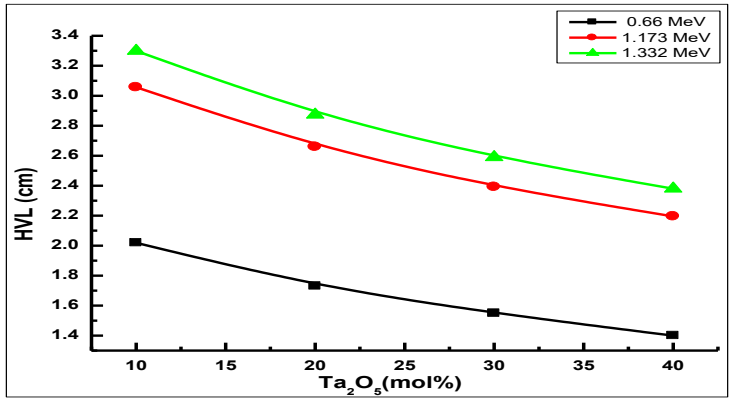


Fig.8: HVL as a function of Ta_2O_5 content for the studied glasses.

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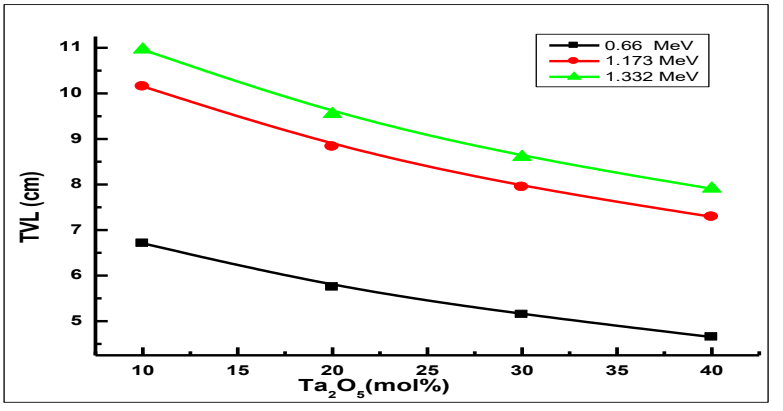


Fig.9: TVL as a function of Ta_2O_5 content for the studied glasses.

Based on the results presented, the BT4 sample exhibited the best shielding ability, as evidenced by its lowest HVL values of 1.40, 2.195, and 2.380 cm and TVL values of 4.651, 7.291, and 7.906 cm at photon energies of 0.662, 1.173, and 1.332 MeV, respectively. In contrast, the BT1 glass sample exhibited the least effective shielding performance, with the highest HVL values of 2.019, 3.057, and 3.299 cm and TVL values of 6.707, 10.15, and 10.95 cm at the corresponding photon energies, respectively. Therefore, the BT4 glass sample can be considered the most promising gamma-ray shielding material among the investigated samples.

MFP is another important quantity to investigation the attenuation of photon and is the reciprocal of μ_L. MFP measures the probability of interaction and represents the average distance traveled by a photon between two successive interactions within a material. As the MFP decreases, the probability of interactions between incoming photons and the atoms of the material increases, leading to greater photon attenuation and absorption [28, 29]. It is worth mentioning that both HVL and MFP are used to estimate the effectiveness of a shielding material. MFP values, as listed in Table 5, fall from 2.91 to 2.02 cm at 0.662 MeV and from 4.76 to 3.43 cm at 1.332 MeV as Ta_2O_5 goes from 10 to 40 mol%. In other words, a photon travels on average about 30% less distance before interacting in TB4 than in TB1. The curves share the shape of the HVL curves (HVL = 0.693 × MFP). Shorter MFP at low energy

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and higher heavy-oxide content is the standard signature of a more effective shield; shorter HVL and MFP with increasing BaO and the lowest HVL at the highest Yb_2O_3 content were reported by previous works [30, 31]. The variation of MFP with photon energy is plotted in Figure 10, while the dependence of MFP on Y_2O_5 content is presented in Figure 13. In general, both figures exhibit the same trend observed for TVL and HVL; thus, MFP variation can be interpreted in the same way as HVL and TVL. These results confirm that the BT4 sample exhibits the smallest HVL, TVL, and MFP values, making it a good candidate for use as a radiation shielding material.

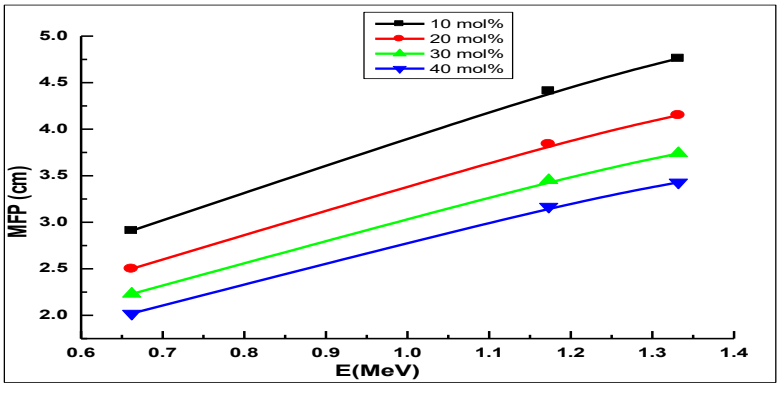


Fig.10: MFP as a function of photon energy for the studied glasses.

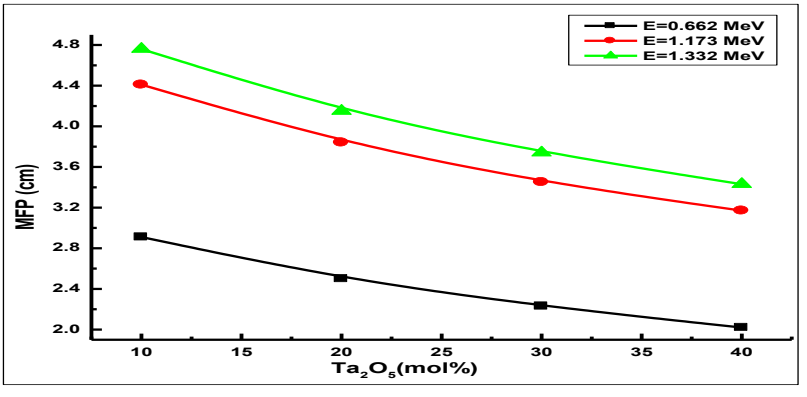


Fig.11: MFP as a function of Ta_2O_5 content for the studied glasses.

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Figure 12 compares the greatest µm value of the current free lead glasses (TB4) with other glasses and concrete at energies of 0.662 MeV, 1.173 MeV, and 1.332 MeV. It is observed that at an energy of 0.662 MeV, the µm value of the glass sample TB4 is relatively better than the values of heavyweight concrete with 36% Pb (HWC36Pb), ordinary concrete (OC), and the free lead glass system ZnOBSi [32 – 34]. Glass TB4 also shows performance comparable to the lead-free glass system 63.8B2O-20K2O-15Bi2O3-0.6Al2O3-0.6CuO (BKBiAlCu) [35]. Furthermore, µm of the glass sample TB4 is slightly lower than that of the glass 60PbO-40B2O3 (PbB), which contains 60% lead [36, 37]. On the other hand, at energies of 1.173MeV and 1.332 MeV, the glass sample TB4 was observed to approach the µm results of the other glass samples and concrete. Based on these results, it is concluded that the lead-free glass sample performs relatively better and is a suitable candidate for radiation protection applications.

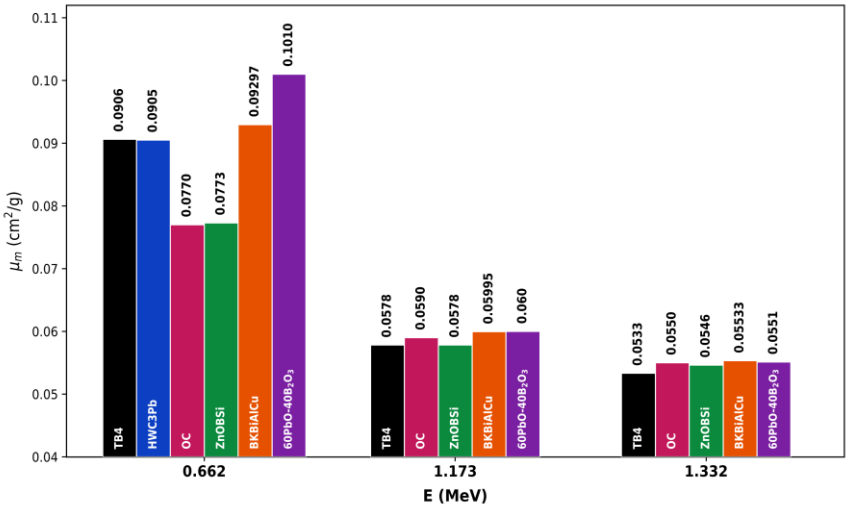


Fig.12: Comparison of the mass attenuation Coefficient of glass samples with some other shielding materials.

4. Conclusions

The influence of Ta_2O_5 content on the physical and shielding properties of the borate glass system, represented by the chemical

formula $xTa_2O_5–20Yb_2O_3–(80–x)B_2O_3$ ($10 \leq x \leq 40$), was systematically investigated. Theoretical estimations of the glass density yielded satisfactory agreement with the expected value. It was observed that increasing Ta_2O_5 concentration led to a rise in both density and molar volume, suggesting an expansion of the glass network and the formation of a more open structural framework within the matrix. The behavior of the oxygen molar volume (V_o) and oxygen packing density (OPD) was contradictory: while V_o exhibited a slight increase, OPD decreased. This indicates that the glass network becomes less tightly packed as the number of bonds within the matrix diminishes. Furthermore, the measured Poisson's ratio corroborates the enhanced compactness of the bonds in the glass matrix upon incorporation of Ta_2O_5 into the network. Moreover, the study's findings explored showed that the incorporation of Ta_2O_5 into yttrium borate glass systems affected certain ionizing radiation shielding properties. Both μm and μL increased with the increment of Ta_2O_5 content because tantalum oxide has a higher absorption capacity for γ rays than B_2O_3. For the case of other parameters (HVL, TVL, MFP), the obtained values were observed to decrease with increasing Ta_2O_5 content because the material becomes more absorptive to radiation, meaning that radiation travels a shorter distance before being absorbed. Overall, the results demonstrate that incorporating Ta_2O_5 into borate glass provides an effective means of modifying its physical properties and radiation-shielding performance, suggesting its potential use in radiation protection applications—particularly glass sample TB4, which exhibited superior performance compared to other glasses and concrete.

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